Source regions and atmospheric deposition of long-range transported Pb in Norway 1977-2000 using moss as indicator

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Lead (Pb) is perhaps the most extensively studied heavy metal in the environment since it was early recognized as toxic to man and biota. This attracted a great interest to learn about sources and behavior of Pb in air, water, and soil, its toxicity and metabolism. Pb is emitted from a variety of emission sources to the atmosphere on fine particles which can be transported very long distances in the air. Therefore Pb pollution is not only a local problem but also significant on a regional and even on a global scale.

Norway has been exposed for centuries to long-range transport of pollution from Central Europe and England but with enhanced amounts of Pb after the onset of adding Pb to gasoline. Various studies of Pb contamination in Norway show a very distinct north – south gradient with excess deposition in the south mainly due to long-range transport of Pb from other parts of Europe. However, regulations of emissions from industry and traffic in many European countries have resulted in a strong reduction of pollutants transported to Norway, as shown e.g. by regular national surveys of heavy metal deposition over the last 25 years based on analysis of moss samples. In the present work contributions from different sources and source regions to the Pb deposited in different parts of Norway over this period are studied by analyzing samples of the moss *Hylocomium splendens* from these surveys with respect to stable Pb isotope ratios.

The ability of mosses to passively accumulate Pb and other metals from dry and wet deposition have made them excellent biomonitors of metal deposition in the environment. Mosses lack a root system and obtain most of their supply of nutrients from precipitation. The absence or strong reduction of the cuticle in these plants means that ions retained on their surface have direct access to exchange sites on the cell wall. Since the thickness of the leaves is only one cell layer, this implies a very close contact with the ambient atmosphere. In this way, nutrients as well as heavy metals are taken up by the mosses. The contact with the underlying mor layer and soil is negligible for most carpet-forming mosses because of weakly developed conductive tissues, and the uptake of heavy metals from the substrate is therefore normally small (Steinnes 1995). Feather mosses growing in natural habitats have therefore become widely used for monitoring of atmospheric deposition of heavy metals.

Since the carpet of living moss tissue is built up during a period of 3-5 years, the content of metals reflects an average exposure during that period. This means that one single analysis of moss may replace years of running sampling and analysis of precipitation, if short-term variations are not of prime interest (Steinnes 1995). The basis of this approach to the study of spatial and temporal deposition patterns is that carpet-forming moss species obtain most of their supply of chemical substances directly from precipitation and from dry deposition of airborne particles. One element for which this technique is particularly suitable is Pb, which is strongly

fixed in the moss and for which the correlation between concentration in moss and bulk deposition is particularly high (Berg et al. 1995).

In this work Pb concentration and Pb isotope ratios were determined by a TIMS in moss samples collected respectively in 1977, 1985, 1990, 1995, and 2000 at 22 sites distributed all over Norway.

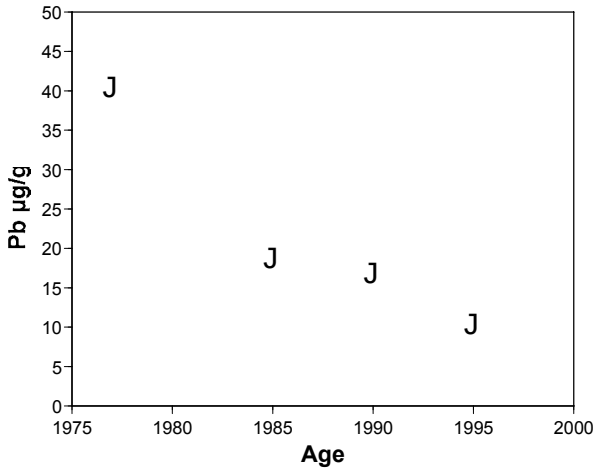

Fig. 1. Average of Pb concentrations for moss samples from Norway, 1977 - 2000.

Fig. 2. Average of 206Pb/207Pb ratios for moss samples from Norway, 1977 - 2000.

The mean values from these analyses confirm the general decrease in Pb concentration (Fig. 1) but also demonstrate a shift in $^{206}Pb/^{207}Pb$ ratios from higher to lower values and then from 1985 and onwards a continuous increase towards Pb from secondary sources (Fig. 2). The average $^{206}Pb/^{207}Pb$ value of European coal from the Czech Republic, Poland and England is about 1.18 (Åberg et al. 1999) while that of European gasoline is about 1.10 with the UK having the lowest ratio of about 1.07 (Kersten et al. 1997).

The $^{206}Pb/^{207}Pb$ ratios thus follow the outphasing of Pb from gasoline starting about 1985. The amount of tetraethyl lead added to gasoline also decreased during this time thus supporting the decrease in total Pb concentration. Part of the large decrease in Pb concentration from 1977 to 1985 can be attributed to the decrease in Pb from coal emission with the closing down on English coal mines and perhaps the beginning of a change to cleaner technologies in the eastern European countries.

In the far north-east of Norway, near the Russian border, the $^{206}Pb/^{207}Pb$ ratios between 1977 and 2000 cluster at about 1.15. A value of 1.15 is also reported by Hopper et al. (1991) and Sturges and Barrie (1989) for aerosols originating from eastern Europe and Russia. Thus the source region for Pb in this region seems to be from the east to southeast. The Pb concentrations in moss samples are generally lower in this region than elsewhere in Norway.

In north-western Norway the $^{206}Pb/^{207}Pb$ ratios vary from less than 1.13 to almost 1.16. The latter values are from year 2000 and the lowest ones from 1985. In this area there are obviously influences from the east with Pb values about 1.15 but also lower values indicating a contribution from south-western and central Europe especially during the 1980s.

Further south, in central Norway, the Pb ratios show a scatter from less than 1.12 to almost 1.16, the lower values from 1985 and the higher from 2000. The impact from south-western and central Europe is more clearly seen here with decreasing isotope ratios from 1977 to 1985 and then slowly increasing towards 2000 due to the outphasing of Pb in gasoline. The Pb concentrations in the moss samples are still low in this region.

In the southwestern part the scatter interval in Pb ratios is the lowest, between 1.11 and 1.15 and the influence of gasoline Pb from England and central Europe is very marked. Over the years there has been a mixing of coal Pb, with a high isotopic ratio, and gasoline Pb, with a low ratio, especially the British type gasoline, rendering the highest Pb concentrations in this area.

In the south-eastern part of the country the isotope pattern is distinctly different from that of the the south-western part, although the lead concentrations in the moss are almost as high. The range of Pb isotope ratios is from about 1.13 to almost 1.17. The impact from England is apparently less here whereas contributions from eastern and central Europe, especially from coal firing, play a greater role.

The geographical distribution of Pb in moss thus shows a gradient of increasing concentration from north to south depending on the Pb input with south and westerly winds to Norway from England and Central Europe. Over time, however, there is a total decrease in Pb concentration over the whole country between 1977 and 2000. An important reason for this is the general fall in the consumption of leaded petrol and coal in western Europe during the last decade, and also the change over to cleaner industrial technologies in the former East European countries.

A comparison with the $^{206}Pb/^{207}Pb$ ratios gives a more clear picture of the evolution and the change from one source to another and shows that it is possible to characterize different sources of pollution. The changes are best seen in southwest Norway where the impact from England, from leaded gasoline with a low $^{206}Pb/^{207}Pb$ ratio, has decreased substancially over the years. Further north the impact from the west is decreasing and substituted by lead pollution from east and southeast, especially in northeast Norway where the Russian impact is very clear.

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